Proton-stopping cross sections and mean excitation energies for gaseous Cl_2 and Br_2

H. Baumgart, H. Berg, E. Huttel, and G. Clausnitzer
Institut für Kernphysik, Strahlenzentrum der Justus-Liebig-Universität Giessen, D-6300 Giessen, Federal Republic of Germany
(Received 22 December 1982)

First measurements of proton stopping powers in gaseous Cl_2 and Br_2 are reported for the energy range between 50 and 750 keV with statistical and systematical errors of about 1% each. In the stopping-power maximum the experimental values are higher than those predicted by the Andersen-Ziegler tables; moreover, lower peak energies are found. Experimental shell corrections were deduced from the proton stopping cross sections and adjusted to the theoretical predictions of Bonderup, whereby higher-order Z_1 correction terms were included. Within this procedure semiempirical values for the mean ionization potentials of 174 eV for Cl_2 and 363 eV for Br_2 were obtained.

The central parameter in the Bethe theory[1] is the mean excitation energy I. Therefore the determination of semiempirical I values from experimental proton stopping cross sections is of basic interest, especially for elements such as Cl_2 and Br_2, where stopping-power measurements were not yet reported.[2] Moreover, these semiempirical values are useful in a comparison with theoretical results which are based on the local-plasma approximation[3] or a single-electron approximation.[4,5]

The electronic stopping cross section of an ion with charge Z_1e penetrating a target with atomic number Z_2 is given for velocities $v > 2Z_1v_0$ (i.e., proton energies above 100 keV) by the Bethe formula as

$$S = \frac{4\pi Z_1^2 e^4}{mv^2} Z_2 L(v,Z_2) \ , \tag{1}$$

with

$$L(v,Z_2) = L_0 = \ln\left(\frac{2mv^2}{I}\right) - ln(1-\beta^2) - \beta^2 - \frac{C}{Z_2} \ , \tag{2}$$

whereby v_0 is the Bohr velocity, m is the electron mass, $-e$ the electron charge, and $\beta = v/c$, with c the speed of light in vacuum. The shell-correction term C/Z_2 is a low-energy correction taking into account the binding of the inner-shell electrons. This term was calculated within the electron-gas energy-loss model by Bonderup.[6] The mean excitation energy is given in the Bethe theory by

$$\ln I = (1/Z_2) \sum_n f_{0n} \ln(\hbar\omega_{0n}) \ , \tag{3}$$

whereby f_{0n} are the dipole oscillator strengths corresponding to the transition frequencies ω_{0n}. Bloch showed[7] that within the Thomas-Fermi model

$$I/Z_2 = \text{const} \ . \tag{4}$$

Chu and Powers[3] calculated I/Z_2 in the framework of the dielectric theory (local-plasma approximation) using Hartree-Fock-Slater atomic charge distributions and found a structure of I/Z_2 vs Z_2, depending on the atomic shell structure. A calculation of I directly with Eq. (3) was performed by Dehmer, Inokuti, and Saxon[4] ($Z_2 \leq 18$) and Inokuti *et al.*[5] ($Z_2 = 19-38$). They evaluated the oscillator-strength distribution from the dipole matrix elements between initial and final states within the independent-electron model using the Hartree-Slater potential. The variation of I/Z_2 with Z_2 is similar to that obtained within the local-plasma approximation whereas the size is about 20% smaller. The possible reasons for this difference were discussed by Inokuti *et al.*

The energy-loss measurements were performed with the same experimental setup and method which was used in a recent experiment with H_2, N_2, O_2, and five inert gases.[8] Now, proton stopping cross sections in Cl_2 and Br_2 were determined for the gaseous phase in the energy range 50–750 keV with a statistical and systematical error of about 1% each. The experimental data are shown in Fig. 1 together with the curves from the Andersen-Ziegler tabulation.[2] It can be seen that the peak positions for the experimental stopping powers are about 20 keV lower than the Andersen-Ziegler values. Furthermore, the Andersen-

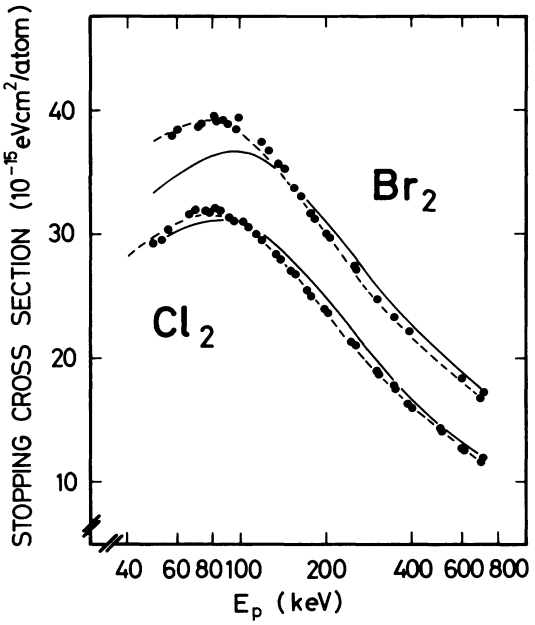

FIG. 1. Proton stopping cross sections in Cl_2 and Br_2. The graph presents our data (●), the values from the Andersen-Ziegler tabulation (full-drawn curves), and a best fit to our data (dashed curves) according to the Andersen-Ziegler semiempirical formula (Ref. 2).

Ziegler curves underestimate the height of the maximum especially for Br_2 ($Z_2=35$). The same trend was recently also found for the adjacent element Kr.[8,9] Since no experimental data exist up to Pd ($Z_2=46$), these results indicate that the Andersen-Ziegler predictions do not represent the stopping-power maximum in a larger Z_2 range.

The procedure used in the following analysis is the one initiated by Besenbacher *et al.*[9] Experimental shell corrections were deduced from the measured stopping powers with use of Eqs. (1) and (2) as

$$\left[\frac{C}{Z_2}\right]'_{\text{expt}} = \ln\left[\frac{2mv^2}{(1-\beta^2)I}\right] - \beta^2 - \frac{1}{Z_2}\frac{mv^2}{4\pi e^4 Z_1^2} S_{\text{expt}} \quad . \quad (5)$$

These are labeled with a dash because inaccuracies of the Bethe theory due to higher-order Z_1 correction terms[10] are not taken into account in this approach. In order to compare the $(C/Z_2)'_{\text{expt}}$ values with the theoretical shell corrections $(C/Z_2)_B$ of Bonderup, corrected values $(C/Z_2)'_B$ including higher-order Z_1 terms were calculated as

$$(C/Z_2)'_B = (C/Z_2)_B - (Z_1 L_1 + L_2) \quad . \quad (6)$$

The Barkas correction L_1 corresponds to a Z_1^3 correction term. Following the suggestion of Lindhard,[10] twice the L_1 term given by Jackson and McCarthy[11] was used. L_2 is the so-called Bloch correction which originates from Bloch's universal stopping formula[12] and corresponds to a Z_1^4 correction in first order.

In Fig. 2, $(C/Z_2)'_{\text{expt}}$ is plotted together with $(C/Z_2)_B$ and $(C/Z_2)'_B$. The $(C/Z_2)'_{\text{expt}}$ values were calculated with the I values of Andersen and Ziegler[2] which were estimated by interpolation between neighboring elements. As can be seen the energy dependence of $(C/Z_2)'_{\text{expt}}$ is well reproduced by $(C/Z_2)'_B$, but there is a shift in the absolute value which may be due to the choice of the I value [see Eq. (5)]. Thus by changing I by a certain amount, the $(C/Z_2)'_{\text{expt}}$ values can be adjusted to the $(C/Z_2)'_B$ curve, whereby a lower I value corresponds to a higher experimental shell correction. With this procedure semiempirical I values were determined from the measured stopping cross sections. These values are given in Table I together with the values of Andersen and Ziegler[2] and theoretical predictions.[3-5] The resulting mean excitation energies are 174 eV for Cl_2 and 363 eV for Br_2 corresponding to I/Z_2 values of 10.24 and 10.37 eV, respectively. These semiempirical I values are both lower than those obtained within the local-plasma approximation,[3] whereas they are higher than those calculated within a single-electron approximation.[4,5]

Some remarks about the reliability of the procedure used for the determination of mean excitation energies should be made. The quality of the determined I values obviously depends on the correctness of Bonderup's theoretical shell corrections and on the higher-order Z_1 corrections to the Bethe theory used in this analysis, i.e., from Lindhard's[10] choice for the Z_1^3 correction, which is twice the magnitude of the earlier calculation of Jackson and McCarthy,[11] and Bloch's Z_1^4 correction of which the Z_1^3 correction term seems to be most critical. If the factor in the Z_1^3 correction should be smaller than 2, this would lead to larger $(C/Z_2)'_B$ values with the consequence that smaller I values would be necessary for the adjustment of the $(C/Z_2)'_{\text{expt}}$ values to the theoretical curves. Therefore the semiempirical I values for Cl_2 and Br_2 given in Table I are certainly upper limits within this model. But on the other hand, there is evidence[13,14] that the higher-order Z_1 correction terms used here (twice the Z_1^3 correction of Jackson and McCarthy combined with the Bloch Z_1^4 correction) are a reasonable choice. In addition, it should be emphasized that the I values for H_2, He, N_2, O_2, Ne, Ar, Kr, and Xe, which were previously determined from proton stopping powers in the same way,[8,9] were in good agreement with those recommended recently

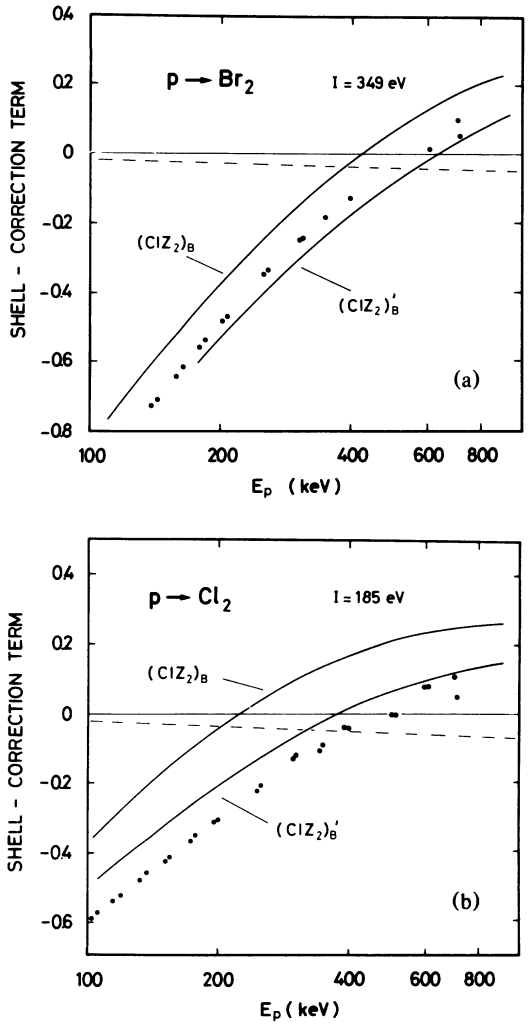

FIG. 2. (a),(b) Experimental shell corrections (●) together with the theoretical values $(C/Z_2)_B$ and $(C/Z_2)'_B$. The $(C/Z_2)'_{\text{expt}}$ values were calculated with the I values given by Andersen and Ziegler (see Table I). The distance between the dot-dashed and the zero line gives the change of $(C/Z_2)'_{\text{expt}}$ due to a 3% change in the stopping-power value.

TABLE I. Mean excitation energies (eV).

	Cl_2	Br_2
Chu and Powers[a]	192	392
Dehmer *et al.*[b]	164	
Inokuti *et al.*[c]		319
Andersen and Ziegler[d]	185	349
This work	174	363

[a]Reference 3; estimated from Fig. 1.
[b]Reference 4.
[c]Reference 5.
[d]Reference 2.

by Ahlen.[15]

Finally, it should be noted that $I = 174$ eV for gaseous Cl_2 is in accord with $I = 173$ eV by Ahlen[15] for liquid Cl. This may be a hint that the gas-solid difference ($I_g/I_s \simeq 0.84$) for Cl_2 proposed in recent stopping-power tabulations[16] is overestimated. It should be pointed out that this was recently also found for the adjacent element Ar.[17]

ACKNOWLEDGMENTS

One of us (H.Ba.) would like to thank Dr. E. Bonderup and Dr. E. Laegsgaard, at the University of Aarhus, for valuable support. This work forms part of the material from his Ph. D. dissertation [University of Griessen, 1983 (unpublished), No. D26].

[1]H. A. Bethe, Ann. Phys. (N.Y.) 5, 325 (1930).
[2]H. H. Andersen and J. F. Ziegler, *Hydrogen Stopping Powers and Ranges in All Elements* (Pergamon, New York, 1977).
[3]W. K. Chu and D. Powers, Phys. Lett. 40A, 23 (1972).
[4]J. L. Dehmer, M. Inokuti, and R. P. Saxon, Phys. Rev. A 12, 102 (1975).
[5]M. Inokuti, J. L. Dehmer, T. Baer, and J. D. Hanson, Phys. Rev. A 23, 95 (1981).
[6]E. Bonderup, K. Dan. Vidensk. Selsk. Mat. Fys. Medd. 35, No. 17 (1967).
[7]F. Bloch, Z. Phys. 81, 363 (1933).
[8]H. Baumgart, W. Arnold, H. Berg, E. Huttel, and G. Clausnitzer, Nucl. Instrum. Methods 204, 597 (1983).
[9]F. Besenbacher, H. H. Anderson, P. Hvelplund, and H. Knudsen, K. Dan. Vidensk. Selsk. Mat. Fys. Medd. 40, No. 3 (1979).
[10]J. Lindhard, Nucl. Instrum. Methods 132, 1 (1976).
[11]J. D. Jackson and R. L. McCarthy, Phys. Rev. B 6, 4131 (1972).
[12]F. Bloch, Ann. Phys. (N.Y.) 16, 285 (1933).
[13]H. H. Andersen, J. F. Bak, H. Knudsen, and B. R. Nielsen, Phys. Rev. A 16, 1929 (1977).
[14]H. H. Andersen, J. F. Bak, H. Knudsen, P. Møller Petersen, and B. R. Nielsen, Nucl. Instrum. Methods 140, 537 (1977).
[15]S. P. Ahlen, Rev. Mod. Phys. 52, 121 (1980).
[16]J. F. Ziegler, *Helium Stopping Powers and Ranges in All Elements* (Pergamon, New York, 1978).
[17]F. Besenbacher, J. Bøttiger, O. Graversen, J. L. Hansen, and H. Sørensen, Nucl. Instrum. Methods 188, 657 (1981).